SU(4) Theory for Spin Systems with Orbital Degeneracy

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Abstract

The isotropic limit of spin systems with orbital degeneracy is shown to have global $SU(4)$ symmetry. On many 2D lattices, the ground state does not posses long range order, which may explain the observed spin liquid properties of $LiNiO_2$. In the $SU(4)$ Neel ordered state, spin-spin correlations can be antiferromagneitc between two neighboring sites with parallel magnetic moments.

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In many transitional metal oxides, the electron configuration on the metal ions has orbital degeneracy in addition to spin degeneracy. In these systems, the sign and magnitude of the spin-spin couplings depends on the orbital occupancy. This may result in interesting magnetic properties of the Mott insulating phase, and is believed to be relevant to unusual properties of many vanadium, titanium, manganese and nickel oxides [1–12]. The Hamiltonian describing spin $s = 1/2$ systems with a two-fold orbital degeneracy (isospin $\tau = 1/2$) was derived by Castellani *et al*. [2]. The Hamiltonian is rotationally symmetric in $\vec{s}$-space, but not in $\vec{\tau}$-space. The aniostropy of the latter is due to Hund's rule and the anisotropy in orbital wavefunctions.

In this Letter we study a simplified Hamiltonian, Eq. (1), where we focus on the rotationally symmetric part in $\vec{\tau}$-space. The insight learned from this higher symmetric model should shed light on our understanding of more realistic systems.

We show the isotropic Hamiltonian (1) has a $SU(4)$ symmetry, and the spin and orbital states are described by four flavors. The simplest $SU(4)$ singlet (flavorless) is a four-site cluster, in analogy to the $SU(2)$ singlet of a two-site pair. The model provides a new possibility for spin liquid ground states in higher dimensions. For the square lattice, using the fermion mean field theory, we find the flavor liquid state to be stable against flavor or generalized spin density wave formation. By comparing the energies of long-ranged ordered states to short-ranged ones on the triangular lattice, we argue the ground state is likely to be a resonant plaquette flavor liquid. In the $SU(4)$ Neel ordered state, the spin-spin correlations can be antiferromagnetic (AF) between two neighboring sites with *parallel* magnetic moments.

The simplest quantum spin-1/2 system with two-fold degenerate orbitals (τ-1/2) and rotational invariance in both $\vec{s}$ and $\vec{\tau}$ -spaces is given by [5]

$$H = J \sum_{\langle i,j \rangle} (2\vec{s}_i \cdot \vec{s}_j + 1/2)(2\vec{\tau}_i \cdot \vec{\tau}_j + 1/2), \tag{1}$$

where $\langle ij \rangle$ is the nearest neighbor (n.n.) pairs. For the AF coupling, which we consider exclusively in this paper, $J > 0$, and we set $J = 1$ hereafter. Apparently, (1) has $SU(2) \times$

$SU(2)$ symmetry, representing rotational invariance in both spin and orbital spaces, and also interchange symmetry between spins and orbitals. We now show that the full symmetry of (1) is actually the higher symmery group $SU(4)$, which unifies the spin and orbital degrees of freedom.

Let us start with an intuitive way to see the $SU(4)$ symmetry by rewriting (1) as,

$$H = (1/4)\sum_{\langle i,j\rangle}(\sum_{\gamma=1}^{15} A_i^\gamma A_j^\gamma + 1), \tag{2}$$

where $A^\gamma = 2s^\alpha, 2\tau^\alpha, 4s^\alpha\tau^\beta$ for $\alpha,\beta = x,y,z$. A^γ can be considered as the fifteen generators of the $SU(4)$ group. We now write them in terms of the more standard generators of group theory.

The Hamiltonian (1) acts on a Hilbert space of 4 basis states at each site. Choosing these as $\mid s^z, \tau^z\rangle$, we label them as

$$\begin{aligned} \mid 1\rangle &= \mid 1/2, 1/2\rangle, \mid 2\rangle = \mid -1/2, 1/2\rangle, \\ \mid 3\rangle &= \mid 1/2, -1/2\rangle, \mid 4\rangle = \mid -1/2, -1/2\rangle. \end{aligned} \tag{3}$$

These basis states form a fundamental representation of $SU(4)$. The conventional $SU(4)$ generators S_m^n acts on a basis state $\mid \mu\rangle$ according to $S_m^n \mid \mu\rangle = \delta_{n,\mu} \mid m\rangle$. The S_m^n's are not hermitian, instead $(S_m^n)^\dagger = S_n^m$. Among the fifteen generators S_m^n, twelve are non-diagonal with six raising and six lowering operators. There is an identity $\sum_{m=1}^4 S_m^m = 1$, so that there are three independent diagonal ones. The S_m^n's are related to $\vec{s}$ and $\vec{\tau}$ by,

$$\begin{aligned} 2s^z &= \sum_{m=1,3}(S_m^m - S_{m+1}^{m+1}), \quad s^+ = \sum_{m=1,3} S_m^{m+1}, \\ 2\tau^z &= \sum_{m=1,2}(S_m^m - S_{m+2}^{m+2}), \quad \tau^+ = \sum_{m=1,2} S_m^{m+2}, \end{aligned} \tag{4}$$

where $s^\pm = s^x \pm is^y$, and $\tau^\pm = \tau^x \pm i\tau^y$. The commutation relations for spin and orbital operators follow from the $SU(4)$ ones, $[S_m^n, S_k^l] = \delta_{n,k}S_m^l - \delta_{m,l}S_k^n$. In terms of S_m^n, (1) becomes

$$H = \sum_{\langle i,j\rangle} S_m^n(i) S_n^m(j), \tag{5}$$

The repeated indices n, m are summed in Eq. (5) and hereafter. We conclude that H has global $SU(4)$ invariance.

Eq. (1), or equivalently (5), gives the effective Hamiltonian for the corresponding Hubbard model in the large U-limit and at 1/4 filling,

$$H_H = -t \sum_{\langle i,j\rangle,\mu} (c^\dagger_{i,\mu}, c_{j,\mu} + h.c.) + U \sum_{i,\mu<\nu} n_{i,\mu} n_{i,\nu}, \tag{6}$$

where $c_{i,\mu}$ and $c^\dagger_{i,\mu}$ are annihilation and creation operators of an electron at site i and state $\mid \mu\rangle$. In terms of electron operators, $S^n_m(i) = c^\dagger_{i,m} c_{i,n}$. Eq. (6) is also equivalent to one of a class of models that has been solved by Sutherland in 1-dimension (1D) [13].

We remark that the AF $SU(4)$ model here is different from the $SU(N)$ model studied by Sachdev and Read [14], and by Arovas and Auerbach [15]. These authors considered AF $SU(N)$ model on bipartite lattices, where the two sublattices have conjugate representations with respect to each other ("quarks" and "antiquarks"). In the present model, all the sites have the same representation, which is not self-conjugate.

To get insight on the physical properties, we first consider systems with a few sites. Since H has global $SU(4)$ invariance, the eigenstates are given by irreducible representations of $SU(4)$. In Fig. 1, we show the Young tableaux for two- and four-site systems. In the two-site system, the lower energy ($\epsilon = -1$) states are 6-fold degenerate (total spin $s = 1$ and total orbital $\tau = 0$ or $s = 0$ and $\tau = 1$), and higher energy ($\epsilon = 0$) states are 10-fold degenerate ($s = \tau = 1$ or both $= 0$). In the four-site system, the ground state is a unique $SU(4)$ singlet $\mid SGL\rangle$, which is rotationally invariant under the SU(4) generators,

$$\sum_i A^\gamma_i \mid SGL\rangle = 0, \tag{7}$$

In terms of S^n_m, the singlet satisfies $\sum_i (S^n_m(i) - \delta_{mn}/4) \mid SGL\rangle = 0$. A $SU(4)$ singlet is a singlet of spin, orbital, and the orbital-spin crossing operator $U^{\alpha,\beta} = 4 s^\alpha \tau^\beta$, and is a generalization of the $SU(2)$ singlet of spin only systems. The energy of the $SU(4)$ singlet of the four-site is found to be $-N_b$, with N_b the number of pairs $\langle ij\rangle$ in (1). Hence, $N_b = 4$ for a 4-site ring, $N_b = 3$ for an open chain, and $N_b = 6$ for a tetrahedron. It is interesting

to note that the energy of each bond in the four-site system is $\epsilon_b = -1$, the best energy a single bond can have. This would be difficult to understand from the conventional valence bond picture for spin only systems, and again indicates the difference between (1) and spin only models including the four-site plaquette RVB state recently discussed in literature [16]. In terms of the fermion operators of (6), the $SU(4)$ singlet can be written as,

$$\mid SGL\rangle = \frac{1}{\sqrt{24}} \sum_{\{ijkl\}} c^\dagger_{i1} c^\dagger_{j2} c^\dagger_{k3} c^\dagger_{l4} \mid 0\rangle, \tag{8}$$

where the sum is over all the permutations of the four sites $ijkl = 1234$.

For large number of sites, N_s, the system can be a $SU(4)$ singlet only if $N_s = 4N$, with N integers. This can be proved easily from (7). A necessary condition for a $SU(4)$ singlet is $\sum_i s^z_i = \sum_i \tau^z_i = \sum_i s^z_i \tau^z_i = 0$. This requires 1/4 of the sites of the system in each of the four flavor states $\mid \mu\rangle$. Systems with $N_s \neq 4N$ may be considered as "edge state" or excitations of $4N$ ones similar to odd-number-site systems in the AF spin-1/2 systems. For an AF $SU(2)$ spin-1/2 chain, there is a theorem [17] that the ground state is a unique spin singlet for finite N_s and, in the thermodynamic limit, is either gapless or has broken translational symmetry (dimerization). Affleck and Lieb generalized this to all 1/2-integer S, and to $SU(N)$ [18]. The $SU(N)$ representations for which their proof applies includes the fundamental representation, which is the representaion for our problem. Thus, the ground state of Eq. (1) for a 1D chain is a unique $SU(4)$ singlet for finite $N_s = 4N$, and in the thermodynamic limit is either gapless or breaks translational invariance ("quadmerization"). Their theorem for $SU(4)$ can be extended to 2D in the same way as for the $SU(2)$ case, but requires a long narrow strip as discussed by Affleck [19].

The $SU(4)$ symmetry identified for model (1) has interesting consequencies. Provided there is no symmetry breaking, it follows from the symmetry that the thermodynamic correlation functions, denoted by $\langle ... \rangle$,

$$\langle s^\alpha_i s^\alpha_j \rangle = \langle \tau^\alpha_i \tau^\alpha_j \rangle = \langle 4 s^\alpha_i \tau^\beta_i s^\alpha_j \tau^\beta_j \rangle = w_{ij}, \tag{9}$$

where w_{ij} is a function of i and j, and is independent of the indices α and $\beta = x, y, z$. This symmetry has been observed in quantum Monte Carlo calculations of the 1D system [20].

For translational invariant systems, the n.n. correlation is related to the energy per bond, ϵ_b,

$$w = \frac{1}{15}(\epsilon_b - \frac{1}{4}), \tag{10}$$

For a 1D chain, Sutherland's Bethe ansatz solution for the equivalent model [13] gives three gapless modes. This may be interpreted in our model as pure spin, pure orbital, and orbital-spin crossing excitations. The correlation function w_{ij} is expected to decay in a power law at T=0, and its sign to be periodic with positive sign if $j - i = 4N$, and negative otherwise. The reason for the latter is the tendency for every four neighboring sites to form a $SU(4)$ singlet.

The nature of the ground state of (1) is of great potential interest. There have been numerous theoretical activities since the discovery of high temperature superconductivity to find possible spin liquid ground states in two or three dimensions. The additional orbital degrees of freedom provides a new possibility for such states. Since there are four *equivalent* single site states $\mid \mu\rangle$ in (1) in comparison with two states in the spin only systems, we expect quantum fluctuations to be stronger, making it more difficult to establish long-range order, hence favoring flavor liquid states.

To illustrate this, we consider model (1) on a square lattice and carry out a fermion mean field theory. In fermion representation, $H = -\sum_{\langle ij\rangle} \chi_{ij}^\dagger \chi_{ij} + constant$, where $\chi_{ij} = \sum_{\mu=1}^4 c_{i,\mu}^\dagger c_{j,\mu}$. The model is similar to the 2D $SU(N)$ t-J model of Affleck and Marston [21], with the important difference that here one fermion per site implies that each flavor of fermions is 1/4 filled, while in their study each flavor of fermions is close to 1/2 filled, the case relevant to cuprates. We consider a uniform and real mean field bond amplitude $\chi = \langle \chi_{ij}\rangle$, and examine its stability against a generalized spin density wave (SDW) state with four sublattices $B_\nu, \nu = 1, 2, 3, 4$. The uniform mean field state describes a flavor liquid. For the spin-1/2 Heisenberg model, the uniform state was found unstable against the (π, π) SDW state [25]. However, the instabilty is related to the nesting Fermi surface at 1/2 filling. We expect the uniform state to become stable against the SDW state at fillings sufficiently

far away from 1/2, as in the present case. Minimizing the mean field energy with respect to the SDW order parameter m, defined so that the mean occupation number for the flavor μ at site $i \subset B_\nu$, $\langle c^\dagger_{i,\mu} c_{i,\mu} \rangle = (1-m)/4 + m\delta_{\nu,\mu}$, we find that the uniform bond state is stable against the SDW. Thus, the mean field theory suggests the ground state of (1) on the square lattice is disordered. The flavor disordered state is found to be gapless in the fermion mean field theory, but from previous studies of $SU(2)$ systems, the issue of the gaplessness needs to be further examined. We note that the 1/4 filled uniform mean field state is unstable against the commensurate flux phase of flux $hc/(4e)$ per plaquette [22,23]. The flux phase is also a flavor liquid.

We expect the model on the triangular lattice to be also disordered. The AF Heisenberg spin-1/2 system on the triangular lattice is believed to order in a three-sublattice 120^0 structure [26]. With orbital degeneracy, such a spin ordering is no longer favored. In Fig. 2, we compare the estimated energies for various long range ordered states, including the classical $SU(4)$ Neel state (the same as the Ising like Neel state for both triangular and square lattices, see reference [24]), the orbital polarized spin ordered state, and a valence bond state, with the $SU(4)$ singlet plaquette solid state. Unlike the spin only problem, where the classical Neel state and the valence bond solid state are degenerate, here the plaquette state has much lower energy than all others. Since the plaquette state can resonate to further lower the energy (and becomes a flavor liquid), we speculate the ground state to be a resonant plaquette state with neither spin nor orbital long range order.

It is interesting to note that a spin liquid state with two-fold orbital degeneracy may have already been realized in the best samples of $LiNiO_2$. The measurements of magnetic susceptibility, specific heat, μSR, and NMR at low temperature show no long range ordering in spins, and the μSR also shows that Ni-spins remain fluctuating even at 20 mK [6]. These evidence strongly indicate a spin liquid ground state. In that material, a formal Ni^{3+}-ion has spin $s = 1/2$ and a two-fold e_g orbital degeneracy. The Ni-ions form layered triangular-lattices, separated by two oxygen and one Li layers, so that the interlayer coupling is weak. We believe the orbital degeneracy is responsible for the observed spin liquid properties, and

Eq. (1) may serve as a simple model to illustrate the role of the orbital degeneracy.

We now turn to the ordered states where the $SU(4)$ symmetry is broken. There are many ways to break $SU(4)$, and here we discuss the generalized 4-sublattice Neel state shown in Fig. 2a. The remaining symmetry is $U(1) \times U(1) \times U(1)$. There are a total of 12 Goldstone modes, comprised of 4 spin-wave modes, 4 orbital-wave modes, and 4 spin-orbital-crossing modes.

The n.n. correlation function can have very unusual properties in states with symmetry breaking. Consider for example the 4-sublattice Neel state. Let the two n.n. sites i and j belong to the two sublattices with $\langle s_i^z \rangle = \langle s_j^z \rangle \geq 0$. In spin only systems, this would imply a ferromagnetic coupling and the correlation $w(s^z) \equiv \langle s_i^z s_j^z \rangle > 0$. In the presence of the orbital degeneracy, the situation can be different. Let us start with the disordered state, where $w(s^z) < 0$ from (10). Provided the transition is continuous, this implies that $w(s^z)$ will remain negative at the transition point, or close to it on the ordered side of the transition. To estimate the critical value of $\langle s_i^z \rangle$, we have carried out calculations on a four-site system. We add a term in (1) representing alternative fields coupled to the spin and orbital, so that the symmetry is explicitly broken, and the four sites represent four sublattices. The ground state and the correlation functions are calculated numerically. We find $w(s^z) < 0$ when $0 < \langle s_i^z \rangle = \langle s_j^z \rangle < 0.18$. Therefore the system can have *antiparallel* spin correlation while both spins have *parallel* magnetic moments. This unusual magnetic property can be tested by neutron scattering.

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FIGURES

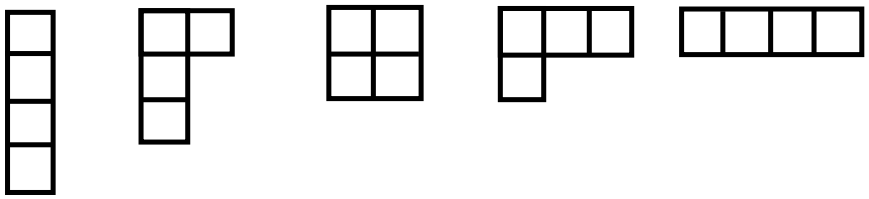
FIG. 1. Young tableaux for $SU(4)$ model (1) a) in a two-site system, and b) in a four-site system. The dimensionality of representations is indicated for each tableau, and inside the parenthesis is the number of distinct representation.

FIG. 2. Average energy per bond ϵ of (1) for various states for the triangular lattice. a) Classical 4-sublattice $SU(4)$ Neel state, with each flavor shown by its orbital (dashed arrow) and spin (solid arrow) state. b) State with orbital "ferromagnetic" and spin AF. In this case, Eq. (1) is reduced to $H = \sum_{\langle i,j \rangle}(2\vec{s}_i \cdot \vec{s}_j + 1/2)$. ϵ is deduced from Ref. [26]. c) Valence bond state. Each double-line represents a two-site valence bond of orbital singlet and spin triplet. Note the spin long range order. d) Plaquette state. Each plaquette (linked by four thick lines) represents a four-site $SU(4)$ singlet.

(a)

6 10

(b)



1 15(3) 20(2) 45(3) 35

Fig.1

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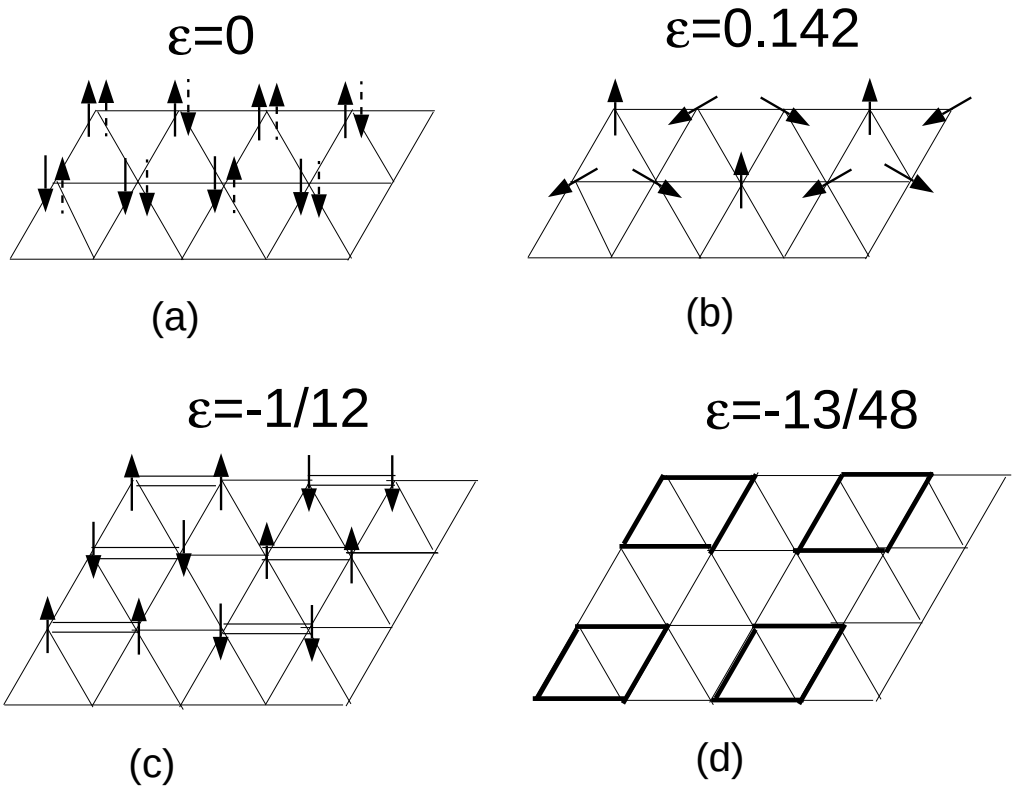

Fig.2

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